Caging dynamics in a granular fluid

P.M. Reis*, R.A. Ingale and M.D. Shattuck
Benjamin Levich Institute, The City College of the City University of New York
140th St. and Convent Av., New York NY 10031, USA

We report an experimental investigation of the caging motion in a uniformly heated granular fluid, for a wide range of filling fractions, ϕ. At low ϕ the classic diffusive behavior of a fluid is observed. However, as ϕ is increased, temporary cages develop and particles become increasingly trapped by their neighbors. We statistically analyze particle trajectories and observe a number of robust features typically associated with dense molecular liquids and colloids. Even though our monodisperse and quasi-2D system is known to not exhibit a glass transition, we still observe many of the precursors usually associated with glassy dynamics. We speculate that this is due to a process of structural arrest provided, in our case, by the presence of crystallization.

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In additional to being of great industrial and geological importance, granular materials are of fundamental interest due to their strong non-equilibrium nature [1]. Ensembles of granular particles are intrinsically dissipative and any dynamical study must involve energy injection. These ingredients make the understanding of granular assemblies a challenging endeavor and a general theoretical framework is still lacking. A possible approach is the study of non-equilibrium steady states, in systems where energy injection perfectly balances dissipation. Whereas some progress has been made in the fast dilute regime [2], the understanding of the dense case of these steady states f remains an open question. One avenue of research has been to borrow concepts from other dense assemblies of particles such as colloids, suspensions and emulsions. Moreover, it is thought that connections with molecular glass formers may be relevant [3]. *A priori*, it is far from obvious that such analogy may be valid due to the enormous differences in lengthscales and the the mechanism of energy supply. However, one common feature in all of these seemingly disparate systems is the presence of *cages*: each particle is temporarily trapped by its neighbors and then moves in short bursts due to nearby cooperative motion. This results in highly heterogeneous motion and slowing down of the dynamics. In molecular systems this behavior is typically observed indirectly from scattering experiments [4]. In colloids, however, caging motion has been observed directly through microscopy, in both 3D [5] and quasi-2D [6] geometries. A large number of theoretical [7, 8] and numerical [9, 10] studies have set out to further investigate the importance of this heterogeneous dynamics. The relevance of the *caging* in driven granular materials [11, 12] and air-fluidized particle systems [13] has only recently started to be addressed. In particular, Dauchot et. al. [12] have reported on a granular system driven by cyclic shear where they observed many "glasslike" features, but for a single value of the filling fraction.

In this Letter we present a novel dynamical study of a monodisperse, quasi-2D granular fluid [14], in which the particles are excited by a spacially uniform stocastic forcing. In our system the filling fraction, ϕ, can be varied from a single particle to hexagonal close packing. We have shown that the structural configurations of this non-equilibrium steady state are the same as those of equilibrium hard-disks [14]. In particular, the system exhibits three phases: an isotropic fluid phase $\phi < \phi_l = 0.652$, a crystaline solid phase $\phi > \phi_s = 0.719$, and an intermediate phase $\phi_l < \phi < \phi_s$ consistent with a hexatic phase[17]. The phase boundaries, ϕ_l (*liquidus* point) and ϕ_s (*solidus point*) are determined by structure alone [14]. Here, we explore the dynamics of these phases through single particle trajectories, focusing on the caging dynamics seen in the intermediate phase. In Fig. 1 we present typical single particle trajectories for filling fractions in each of the three phases. Simple fluid behavior is observed at low ϕ, characterized by random diffusion (Fig. 1a). Above crystallization ($\phi > \phi_s$) particles

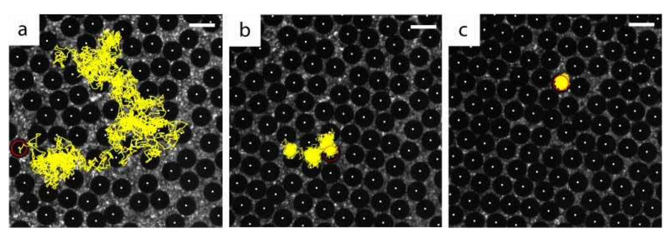

FIG. 1: Experimental frames with superposed typical trajectories of a single particle: (a) $\phi = 0.567$, (b) $\phi = 0.701$ and (c) $\phi = 0.749$. Note that even though only a single trajectory is shown for each ϕ, particle tracking and statistics were performed over all particles within the imaging window. The scalebar is 2mm.

*Current address: Laboratoire PMMH (UMR7636 CNRS-ESPCI-P6-P7), 10 rue Vauquelin, 75231 Paris, FRANCE. preis@pmmh.espci.fr

become fully arrested by thier six hexagonally packed neighbors (Fig. 1c). In the intermediate phase, we see a mixture of these behaviors (Fig. 1b). At short times, particles are temporarily trapped in cages formed by thier neighbors, but at long times they diffuse from cage to cage. We will use the Mean Square Displacement (MSD) and the Intermediate Scattering Function (ISF) to show that the caging dynamics seen here is qualitatively identical to that of dense molecular and colloidal systems [4, 5] and supercooled liquids [15]. This is surprising since our experiment is fundamentally different. The associated lengthscales of our granular system are typically 3-7 orders of magnitude larger than those of molecular systems and colloids. Moreover, the steady states we study are inherently far from equilibrium since energy is both injected (through vibration) and dissipated in inter-particle inelastic collisions and frictional contacts.

Our experimental apparatus consists of vertically vibrated, $D = 1.19mm$ diameter stainless steel spheres confined between two $85.3D$ diameter horizontal glass plates, separtated by $1.6D$, which constrain the particle motion to be quasi-2D. Our system is described in more detail in [14, 16] and improves on Olafsen and Urbach's similar system [19] by using a roughned bottom plate. This allows us to study a wide range of filling fractions ($1.4 \times 10^{-4} < \phi < 0.8$). We sinusoidally vibrate the system with a frequency $f = 50Hz$ and a maximum acceleration equal to 4 times gravity, but the structure and dynamics of the system are independent of the forcing for a wide range of parameters [16]. To ensure repeatable initial conditions, we start with all particles hexagonally packed near the boundary. A steady-state is reached by waiting for 12000 vibration cycles before 10 seconds of data are acquired. We record the dynamics in a ($15 \times 15mm^2$) central region using a high-speed camera at 840 Hz and track the trajectories of all particles.

We first analyse the particle trajectories by measuring the Mean Square Displacement defined as,

$$M(t) = \langle [r(t) - r(0)]^2 \rangle, \quad (1)$$

where $r(t)$ is the position of a particle at time t, $r(0)$ is its initial position, the brackets $\langle\rangle$ signify ensemble averaging over many realizations and time invariance is assumed. The MSD for a range of ϕ are shown in Fig. 2. For the case of a single particle in the cell (marked with a square in Fig. 2), the motion at short times is ballistic, and $M(t) \sim t^{\alpha}$ where $\alpha \sim 2$ ($\alpha = 2$ for pure ballistic motion). At later times, the particle moves diffusively and the slope of $M(t)$ tends to $\alpha = 1$. This shows that the trajectory of a single particle is indeed randomized across the cell. For all ϕ, the motion at early times is superdiffusive with $\alpha \sim 2$ showing ballistic motion. For $\phi < 0.719$ the motion always becomes diffusive at long times with $M(t) \sim t$. Eventually, this becomes increasingly noisy due to the lack of statistics in the time averaging. For $\phi > 0.719$ the particles are trapped by their six hexagonally packed neighbors, and $M(t)$ levels off to a constant value set by the lattice spacing. In the intermediate phase, however, a plateau emerges at intermediate times where the motion is subdiffusive with $0 < \alpha < 1$. This plateau becomes obvious above the liquidus point, ϕ_l (marked as * in Fig. 2) and represents the slowing down due to the cage effect shown in Fig. 1b. A similar dependence of $M(t)$ has recently been observed in a quasi-2D system of bidispersed particles fluidized by a uniform upflow or air [13].

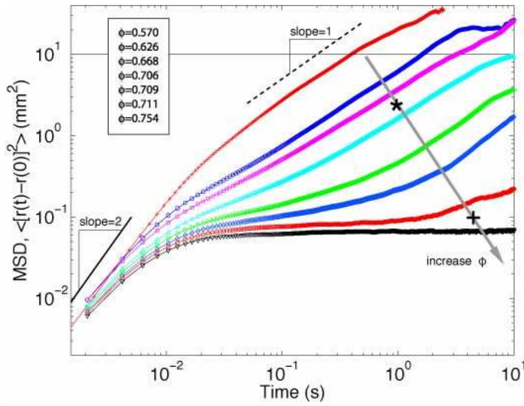

FIG. 2: Time dependence of the Mean Square Displacement for various values of filling fraction (numerical values given in the box). The curve marked with a square is for a single particle in the cell. The arrow points in the direction of increasing ϕ. Along the arrow, the symbols (*) and (+) are located at ϕ_l and ϕ_s, respectively. The horizontal line corresponds to the area of 1/4th of the system, above which finite system size effects become important.

Another classic measure in the study of dense liquid phases is the Intermediate Scattering Function [4] which is defined as,

$$F_s(\mathbf{q}, t) = \frac{1}{N} \sum_j \langle \exp\left(-i\mathbf{q} \cdot [\mathbf{r}_j(t) - \mathbf{r}_j(0)]\right) \rangle, \quad (2)$$

where $\mathbf{q}$ is a wavenumber and $\mathbf{r}_j(t)$ is the trajectory of particle j out of N particles in the system. This measure is widely used in colloids since it is readily available through light scattering experiments [4] and is, essentially, a measure of the time decorrelation of the positional wavevectors. In dense colloids and supercooled liquids, $F_s(t, q)$ captures the relaxation due to caging in the form of a two-step relaxation: 1) the fast (early time) β relaxation which corresponds to the diffusion inside the cage followed by 2) the α relaxation corresponding to the time it takes for the particle to diffuse out of the cage.

In Fig. 3 we plot $F_s(t)$ for wavevector $qD = 2.14$ ($q = 1.8mm^{-1}$) for various values of ϕ. As expected, in the crystal phase ($\phi > 0.719$) F_s levels at a value close to 1 and little decorrelation occurs, since each particle is fully trapped. In the fluid phase, F_s rapidly decays as

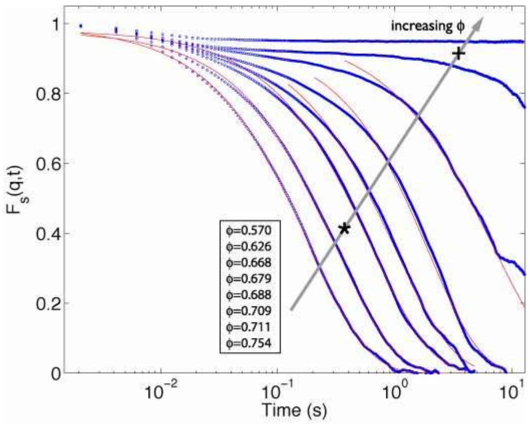

FIG. 3: Time dependence of the Intermediate Scattering Function, with $qD = 2.14$, for various filling fractions (numerical values of ϕ given in the box). The arrow points in the direction of increasing ϕ. Along the arrow, the symbols (*) and (+) are located at ϕ_l and ϕ_s, respectively. The solid lines are fits to Eqn. (3).

particles move across the cell diffusively and the initial positional wavevector quickly decorrelates. In the intermediate phase we observe the classic α and β two-step relaxation; there is a clear intermediate plateau and the α relaxation occurs at increasingly longer timescales, as ϕ is increased. As for the MSD before, this two-step relaxation becomes visible above the liquidus point, ϕ_l (marked as * in Fig. 3). For each value of ϕ, the α relaxation at late times is well described by a stretched exponential of the form,

$$F_s(q,t) \sim \exp[-\left(t/\tau(q)\right)^{\beta(q)}], \tag{3}$$

where $\tau(q)$ is a relaxation time and the stretching exponent is typically $\beta(q) \leq 1$. Fits of this stretched exponential form to the experimental data are shown as solid lines in Fig. 3. It is interesting to note that this behavior is in good agreement with the predictions of Mode Coupling Theory (MCT) [7]. This is highly surprising since MCT has been developed for thermal fluids and is not known to apply to non-equilibrium systems such as ours.

We now focus on the q-dependence of both the relaxation time $\tau(q)$ (Fig. 4a) and the exponent $\beta(q)$ (Fig. 4b), for various values of ϕ. We defined $\tau(q)$ as the time it takes for the experimental curves of F_s to fall to a level of $1/e$, i.e. $F_s(\tau) = 1/e$. The stretching exponent $\beta(q)$ is the local slope of the quantity $\log(-\log(F_s))$, in the neighborhood of τ. At low filling fractions, τ scales as q^{-2}. As the filling fraction is increased this scaling continues to hold but only up to a cut off value above which (lengthscale below which) it sharply drops. This cutoff lengthscale can be associated with a characteristic size of the cage, which becomes increasingly smaller as ϕ is increased. On the other hand β, the local exponent, tends to one at small q but decreases progressively below one for higher filling fractions and F_s becomes increasingly stretched. These findings can now be combined and interpreted as follows. For small q (i.e. large lengthscales), $\tau \sim q^{-2}$ and $\beta(q) \to 1$, which together imply that $F_s(q,t) \sim \exp(-Dq^2t)$. This is the result for classical diffusion [12]. For large q (i.e. small lengthscales) this Brownian scaling breaks down to a stretched exponential with $\beta < 1$, which can attributed to the presence of dynamic heterogeneities due to caging.

Returning to the case of fixed $qD = 2.14$, we plot τ as a function of ϕ in Fig. 5. A significant slowing down of the dynamics can be seen at high ϕ, as crystallization is approached. It is highly surprising that this slowing down with ϕ is well described by the Vogel-Fulcher law [20] as found in many glass forming systems,

$$\tau \sim \exp[A/(\phi_c - \phi)], \tag{4}$$

where $A = 0.094 \pm 0.004$ is a fitting parameter but $\phi_c = 0.719 \pm 0.007$ is the filling fraction for crystallization which was determined independently from experiments [14]. A power-law fit was not as satisfactory. Note that, as the fluid goes through the transition from isotropic fluid to the intermediate phase (at $\phi_l = 0.652$), τ shows no particular feature. This functional dependence of the

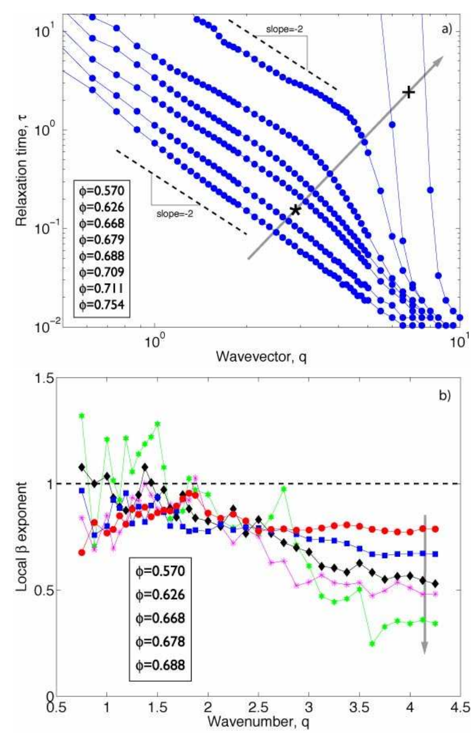

FIG. 4: (a) Wavevector dependence of the relaxation time, τ and (b) local stretching exponent β, for various values of filling fraction. The arrows point in the direction of increasing ϕ and the numerical values of ϕ are given in the boxes. Along the arrow, the symbols (*) and (+) are located at ϕ_l and ϕ_s, respectively.

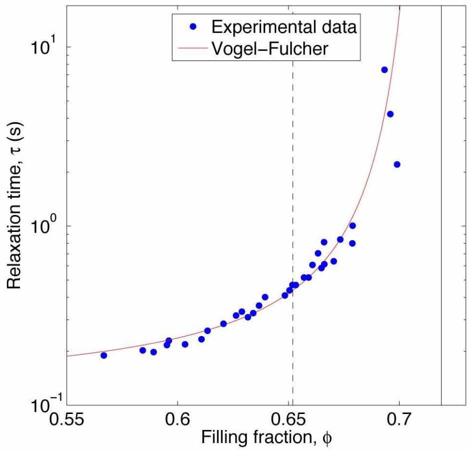

FIG. 5: Relaxation time, τ, extracted from the Intermediate Scattering Function as a function of filling fraction. The solid line is a fit to the Vogel-Fulcher law of Eqn. (4). Dashed and solid lines represent the location of the liquidus and solidus points, respectively.

relaxation has also recently been found within the granular context by Fierro *et. al.* [21] in a numerical lattice model.

In summary, we have studied the dynamics of a uniformly heated granular fluid. We have observed a number of features typically associated with dense liquid behavior in molecular and colloidal systems, namely: prominence of cages, development of a plateau in the MSD and ISF with the breakdown of the Brownian diffusive behavior and a Vogel-Fulcher relaxation. These features become particularly visible above the liquidus point, for $\phi \geq \phi_l$. In particular, our results can be directly compared to both experiments [6] and simulations [10] of quasi-2D colloidal particles. This is surprising since our experiment is fundamentally different: it is intrinsically far from equilibrium since energy is not conserved, the constituent particles are macroscopic and it is known that for monodisperse and quasi-2D systems such as ours there is no ideal glass transition [18] (the phase diagram is fluid, intermediate phase and crystal). In the absence of a structural glass, we propose that the spatially uniform stochastic way of injecting energy along with a process of structural arrest provided by crystallization [14] are at play to account for the many observed similarities. This suggests that theoretical frameworks previously developed for dense thermal liquids, for example MCT [7], might shed some light to the description of excited granular materials.

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